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SRL  
RECORD COPYCALCULATION OF THREE-DIMENSIONAL POLLUTANT  
TRANSPORT USING A METHOD-OF-MOMENTS TECHNIQUE

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## INTRODUCTION

Accurate prediction of the transport of radioactive and/or non-radioactive pollutants released into the environment is required to assess potential hazards to the public. Environmental modeling of such releases is necessary in atmospheric as well as estuary, river, stream, and ground water flows. Only under ideal conditions can the dispersion of concentration from a source be accurately calculated using analytical methods. However, such methods are typically insufficiently flexible to handle complex cases of three-dimensional time-dependent dispersion when environmental conditions are constantly changing.

Numerical models, while more flexible in solving complex transport problems, generally suffer from numerical errors. Herein, a three-dimensional method-of-moments technique is employed to calculate pollutant advection. The method is specifically derived to minimize numerical diffusion and dispersion error, and is based on the calculation of moment distributions of a concentration within a cell, cf. Egan and Mahoney (1972), Pedersen and Prahm (1974), Fischer (1977), Pepper and Long (1978). By summing the moments over the entire solution domain, and using a Lagrangian advection scheme, a concentration field can be transported without generation of numerical dispersion error. Because the method maintains subgrid scale resolution, point and area source releases can be calculated without significant computational damping. To reduce computer programming complexity and computation time, the time-splitting procedure of fractional steps, Yanenko (1971), is used to calculate the three-dimensional solutions. Three-dimensional diffusion is

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solved by the method of cubic splines (Price and MacPherson, 1973; Rubin and Graves, 1975; Ahlberg, et al., 1967; Fyfe, 1969). The cubic spline method is based on continuous-curvature cubic spline relations used as interpolation functions for first and second derivative terms. The method of fractional steps is likewise used as in the advection solution procedure. After solution of the diffusion terms, the first and second moments are recalculated to insure continuity with the advection terms.

Results are presented on the ground level concentrations of tritium calculated with the three-dimensional model. Results are compared with experimental data obtained after the accidental release of tritium on May 2, 1974 from Savannah River Plant. The effect of topography on plume emissions is also examined, under ideal conditions and results compared with values obtained from the conventional Gaussian plume relations.

#### PROBLEM STATEMENT

The governing equation for three-dimensional time-dependent pollutant transport can be written as

$$\frac{\partial C}{\partial t} + \vec{U} \cdot \nabla C = \nabla \cdot (\vec{K} \nabla C) + S \quad (1)$$

where  $C$  is concentration ( $gm/m^3$ ),  $\vec{U}$  is the vector velocity field ( $m/sec$ ),  $\vec{K}$  is the directionally-dependent eddy diffusivity (exchange coefficient of diffusion,  $m^2/sec$ ), and  $S$  represents the source and sink terms associated with precipitation, scavenging, source emission, deposition and chemical reaction. Equation (1) is solved within the physical region,  $x_W < x < x_E$ ,  $y_S < y < y_N$ ,  $h(x,y) < H$  for  $t > 0$ , where  $x_W$ ,  $x_E$ ,  $y_S$ ,  $y_N$  are the west, east, south, and north lateral boundaries within the  $x$ - $y$  plane,  $h(x,y)$  is the ground elevation at  $(x,y)$ , and  $H$  is the elevation of the upper limit for vertical mixing (atmospheric lid). An analogous equation which includes a variable mixing height,  $H(x,y,t)$ , is used by Reynolds, et al. (1973). Since this study is concerned with releases which occur only over a few hours, the variation of the mixing height is assumed to be negligible.

The initial condition on Equation (1) is that the mean concentration is everywhere zero.

$$C(x,y,z,t=0) = C_0 = 0 \quad (2)$$

The mean concentration could also be specified at all locations as some non-zero initial background value. The following set of boundary conditions are used to constrain the solution to the domain of interest:

$$\underline{z = h(x,y):}$$

$$-\hat{K} \nabla C_0 \cdot \hat{n}_h = f_0 \quad (3)$$

$$\underline{z = H:}$$

$$-\hat{K} \nabla C_0 \cdot \hat{n}_H = 0 \quad (4)$$

$$\underline{x = x_W, x_E \text{ and } y = y_N, y_S:}$$

$$-\hat{K} \cdot \nabla C_0 \cdot \hat{n} = 0 \text{ for } \vec{U} \cdot \hat{n} > 0 \quad (5)$$

$$(\vec{U} C_0 - \hat{K} \nabla C_0) \cdot \hat{n} = \vec{U} C_0 \cdot \hat{n} \text{ for } \vec{U} \cdot \hat{n} < 0 \quad (6)$$

where  $\hat{K}$  is the diagonal matrix of the eddy diffusivity tensor,  $f_0$  is the mass flux of concentration at the surface (for puffs or plumes  $f_0 \equiv 0$ ),  $\hat{n}_h$  is the unit vector normal to the surface while  $\hat{n}_H$  is the outward directed unit vector normal to the surface defined by the inversion base. Also,  $\hat{n}$  is the outward directed unit vector normal to the horizontal boundary, and  $C_0$  is the mean concentration just outside the solution domain.

To account for deposition at the surface, the flux at the ground is expressed in terms of a deposition velocity, Calder (1963), as

$$pC \approx V_g C + \left( K_z \frac{\partial C}{\partial z} \right)_{z=0} \approx (1-r) K_z \left( \frac{\partial C}{\partial z} \right) \quad (7)$$

where  $V_g$  is the actual settling velocity,  $p$  is the deposition velocity, and  $r$  is the reflection coefficient. Ranging  $r$  from 0 to 1 simulates the effect of losses at the surface by deposition.

The non-regularity of the closure surface of the three-dimensional modeled region is removed by the coordinate transformation, cf. Reynolds et al. (1973),

$$\xi = \frac{x - x_E}{X}, \quad \underline{X} = x_W - x_E \quad (8)$$

$$\eta = \frac{y - y_S}{Y}, \quad \underline{Y} = y_N - y_S \quad (9)$$

$$\rho = \frac{z - h(x,y)}{Z}, \quad \underline{Z} = H - h(x,y) \quad (10)$$

After some algebra, Equation 1 becomes

$$\begin{aligned}
 \frac{\partial C}{\partial t} + \frac{U}{X} \frac{\partial C}{\partial \xi} + \frac{V}{Y} \frac{\partial C}{\partial \eta} + \frac{\tilde{W}}{Z} \frac{\partial C}{\partial \rho} = & \frac{1}{X^2} \frac{\partial}{\partial \xi} \left[ K_x \left( \frac{\partial C}{\partial \xi} - \Lambda_\xi \frac{\partial C}{\partial \rho} \right) \right] \\
 & - \frac{\Lambda_\xi}{X} \frac{\partial}{\partial \rho} \left[ \frac{K_x}{X} \left( \frac{\partial C}{\partial \xi} - \Lambda_\xi \frac{\partial C}{\partial \rho} \right) \right] \\
 & + \frac{1}{Y^2} \frac{\partial}{\partial \eta} \left[ K_y \left( \frac{\partial C}{\partial \eta} - \Lambda_\eta \frac{\partial C}{\partial \rho} \right) \right] \\
 & - \frac{\Lambda_\eta}{Y} \frac{\partial}{\partial \rho} \left[ \frac{K_y}{Y} \left( \frac{\partial C}{\partial \eta} - \Lambda_\eta \frac{\partial C}{\partial \rho} \right) \right] \\
 & + \frac{1}{Z^2} \frac{\partial}{\partial \rho} \left[ K_z \frac{\partial C}{\partial \rho} \right] + S \cdot Z \quad (11)
 \end{aligned}$$

where

$$\Lambda_\xi = \frac{1}{Z} \left( \frac{\partial h}{\partial \xi} + \rho \frac{\partial Z}{\partial \xi} \right) \quad (12)$$

$$\Lambda_\eta = \frac{1}{Z} \left( \frac{\partial h}{\partial \eta} + \rho \frac{\partial Z}{\partial \eta} \right) \quad (13)$$

$$\tilde{W} = W - \frac{U}{X} \Lambda_\xi Z - \frac{V}{Y} \Lambda_\eta Z \quad (14)$$

A three-dimensional wind field and diffusion coefficient distribution is required established for solution of Equations (11)-(14). The mean velocity vector is assumed parallel to the ground and lid, i.e.,  $U \neq 0$ ,  $V \neq 0$ , and  $W = 0$ .

In order to specify the wind field throughout the three-dimensional region, a subjective analysis and interpolation scheme is used to calculate a first guess wind field based on available data, i.e., wind speeds and directions obtained from instrumented towers. The three-dimensional wind field is initially constructed from interpolation of experimental data. A mass consistent wind field model is then used to calculate corrections to the interpolated wind vectors at each node point such that continuity is satisfied, i.e.,

$$\frac{\partial U'}{\partial \xi} + \frac{\partial V'}{\partial \eta} + \frac{\partial \tilde{W}}{\partial \rho} = 0 \quad (15)$$

where  $\tilde{W}$  is given by Equation (14),  $U' = UZ$  and  $V' = VZ$ . Based on the techniques of Dickerson (1975) and Sherman (1977), a Sasaki variational statement is minimized to determine the mass-consistent correction to the initial velocity field.

Based on Yu's (1977) analysis of 14 different models for determining the vertical diffusion coefficient, an O'Brien (1970) K-theory model is used in conjunction with similarity theory to determine the diffusion coefficient distribution. Surface similarity theory is used to calculate  $K_z$  from the Monin-Obukhov universal relations with measured wind velocities and temperatures from an instrumented TV tower within the transition layer region ( $z \sim 60$  m). The O'Brien cubic profile is then used to calculate the vertical diffusivity above the transition layer region to the top of the mixing layer. This procedure was used by Pepper and Kern (1978) to model atmospheric dispersion with linear finite element and cubic spline methods. For the present calculations,  $K_x \equiv K_y$ .

#### THE NUMERICAL MODEL

To reduce computer core requirements for solution of three-dimensional concentration transport, Equation (11) is split into a series of one-dimensional equations by the method of fractional steps, Yanenko (1971). Equation (11) is divided as:

$$\frac{\partial C^*}{\partial t} + \frac{U}{X} \frac{\partial C^*}{\partial \xi} + \frac{V}{Y} \frac{\partial C^*}{\partial \eta} + \frac{\tilde{W}}{Z} \frac{\partial C^*}{\partial \rho} = 0 \quad (16)$$

$$\begin{aligned} \frac{\partial C^{**}}{\partial t} = & - \frac{1}{X^2} \frac{\partial}{\partial \xi} \left[ K_x \Lambda_\xi \frac{\partial C^*}{\partial \rho} \right] - \frac{\Lambda_\xi}{X} \frac{\partial}{\partial \rho} \left[ \frac{K_x}{X} \frac{\partial C^*}{\partial \xi} \right] \\ & - \frac{1}{Y^2} \frac{\partial}{\partial \eta} \left[ K_y \Lambda_\eta \frac{\partial C^*}{\partial \rho} \right] - \frac{\Lambda_\eta}{Y} \frac{\partial}{\partial \rho} \left[ \frac{K_y}{Y} \frac{\partial C^*}{\partial \eta} \right] \end{aligned} \quad (17)$$

$$\frac{\partial C^{***}}{\partial t} = \frac{\Lambda_\xi}{X} \frac{\partial}{\partial \rho} \left[ \frac{K_x \cdot \Lambda_\xi}{X} \frac{\partial C^{**}}{\partial \rho} \right] + \frac{\Lambda_\eta}{Y} \frac{\partial}{\partial \rho} \left[ \frac{K_y \cdot \Lambda_\eta}{Y} \frac{\partial C^{**}}{\partial \rho} \right] \quad (18)$$

$$\begin{aligned} \frac{\partial C^{n+1}}{\partial t} = & \frac{1}{\underline{x}^2} \frac{\partial}{\partial \xi} \left[ K_x \frac{\partial C^{***}}{\partial \xi} \right] + \frac{1}{\underline{y}^2} \frac{\partial}{\partial \eta} \left[ K_y \frac{\partial C^{***}}{\partial \eta} \right] \\ & + \frac{1}{\underline{z}^2} \frac{\partial}{\partial \rho} \left[ K_z \frac{\partial C^{***}}{\partial \rho} \right] \end{aligned} \quad (19)$$

Successive solutions to Equations (16) through (19) give the final solution to Equation (11) at one time step. The Equation set (16)-(19) can be reduced to only Equations (16) and (19) when changes in ground elevation are gradual. Then, derivatives  $\partial h / \partial \xi$ ,  $\partial h / \partial \eta$ ,  $\partial Z / \partial \xi$ , and  $\partial Z / \partial \eta$  are negligible compared to other terms in the equation set. Hence, the diffusion terms containing  $\Lambda_{\xi}$  and  $\Lambda_{\eta}$ , Equations 17 and 18, are neglected.

The method of second moments (Egan and Mahoney, 1972) is used to solve Equation (16). The method calculates the zeroth, first, and second moments of the concentration within a mesh and then advects and diffuses the concentration by maintaining conservation of the moments. The moments correspond to the mean concentration, center of mass, and scaled distribution variance (moment of inertia), respectively, and are given by

$$C_m = \int_{-0.5}^{0.5} C(\xi_m) d\xi \quad (20)$$

$$F_m = \int_{-0.5}^{0.5} C(\xi_m) \xi_m d\xi / C_m \quad (21)$$

$$R_m^2 = 12 \int_{-0.5}^{0.5} C(\xi_m) (\xi_m - F_m)^2 d\xi / C_m \quad (22)$$

where  $\xi_m$  denotes the relative displacement of material within the  $m^{\text{th}}$  cell from the center of the cell.  $\xi_m$  varies from -0.5 to +0.5 corresponding to the left and right hand extreme boundaries of a cell. For a simple rectangular mesh, the integrals are evaluated by summation for each grid element in terms of the concentration distributions of the portions remaining and newly transported in for each successive time step. For illustration, the transport of a single cell of concentration is shown in Figure 1 for two-dimensional advection. Note the single cell is advected without numerical dispersion or computational damping errors. Similar tests on hyperbolic equations with both finite difference and finite

element techniques are discussed by Long and Pepper (1976) and Baker, et al. (1978). Equations (16)-(19) are recast as algebraic equation systems using cubic spline interpolation to establish derivatives, cf. Rubin and Graves (1975). Complete details of the algorithm are given by Pepper and Baker (1978).

## NUMERICAL RESULTS

The computational domain normally consists of 10,890 cells; 33 cells in the longitudinal direction ( $\xi$ ), 33 cells in the lateral direction ( $\eta$ ), and 10 levels in the vertical direction ( $\rho$ ). Mesh spacing can either be arbitrarily set (such as a telescoping grid network) or equally spaced with  $\Delta\xi = \Delta\eta$ . The vertical spacing is based on the ground level values for topography and the height of the lid. User input values for the remaining levels, i.e., levels corresponding to instrumented tower locations, are automatically transformed to non-dimensional values such that  $0 < \rho < 1$  throughout the computational domain.

To assess model accuracy for advection of a continuous area source, a simple test was conducted using a six cell source, each with a unit release advected in a two dimensional constant wind field. Figure 2 compares numerical predictions to an analytical solution (Pedersen and Prahm, 1974). The results are nearly identical, with computed peak centerline values as well as the width of the plume accurately maintained. All remaining values in the computational domain are zero, in contrast to predictions using standard finite difference procedures which tend to produce wider plume width and associated loss of centerline concentration. The numerical values are identical to those of Pedersen and Prahm (1974) obtained using a two dimensional method of moments technique. In both cases a width correction procedure has been used to eliminate small lateral dispersion at the plume edge, see Pepper and Long (1978). Additional test case comparisons are reported in Pepper and Baker (1978).

The effect of surface irregularity on prediction of dispersion of a continuous elevated emission has been evaluated. Figure 3 shows the elevation distribution of the ground plane; the continuous release was assumed to occur at a height of 250 m at the left-center cell denoted with a dot. A 200 m peak surface elevation occurs 11 km downwind. The height of the lid is constant at 650 m. Equal grid spacing was used in the x-y plane, spanning a 400 m<sup>2</sup> area, with  $\Delta X = \Delta Y = 1000$  m; vertical grid spacing was equally incremented in 100 m intervals. The source continuous emission rate was set at 1 gm/sec. Atmospheric stability conditions are assumed neutral. The transport coefficients were  $K_y = 33$  m<sup>2</sup>/sec throughout the domain and  $K_z = f(z)$ , obtained from Pasquill stability curves at 1000 m distances, see Slade (1968), Chapter 3. The initial velocity field is given as



$$U = 5.0 \left( \frac{Z}{.2} \right)^{.14}$$

$$V = W = 0$$

Hence, at the source elevation of 200 m, the velocity field is  $\vec{U}_{200} = (13.15, 0, 0)$  m/sec.

Concentration isopleths and the computed mass-consistent vector wind field are shown in Figure 4 in the mid-plane of the computational domain. In Figure 4.a, the vector denotes the magnitude of the wind speed and the vertical scale has been increased to enhance visualization of the small vertical velocities. The wind field was held constant, and the essential steady-state concentration isopleths are shown in Figure 4.b. For reference, Figure 4.c illustrates topography, and maximum concentrations are observed clustered around the peak elevation. The effect of topography on the vertical distribution of concentration is shown in Figure 5. Ground level centerplane C/Q values are plotted as a function of longitudinal distance downwind, and the influence of topography is substantial. Computed solution trends for both cases are in agreement with a steady-state Gaussian plume analytical solution (adjusted for topography, see Kao, 1976).

The final test case corresponds to prediction of an actual release of tritium from the Savannah River Plant (SRP) for which experimental data is available. Averaged over the four-minute release period, tritium was present in the exhaust air at a concentration of approximately 13 ppm by volume. Because of this low dilution, the discharge was assumed to behave as a neutral gas with no buoyancy. At the release, a polar front was located approximately 80 km north of SRP, see Figure 6. This front represents a dividing line between two different wind fields present in the Carolinas during and after the release. Winds were predominantly from the southwest below the front. Sky conditions were generally overcast with precipitation north of the front.

The vector wind field and initial concentration distribution at the 62-m height over SRP are shown in Figure 7. The overcast skies and generally steady winds produced atmospheric diffusion corresponding to Pasquill Category D -- slightly stable. Measurements obtained with an acoustic sounder indicated that the height of the mixed layer was approximately 610 m. Hence, the concentration puff was assumed to be transported and diffused by the wind field below this height. The solution was initiated with the source term set equal to  $4.79 \times 10^5$  Ci. Wind field data were obtained from the National Weather Service and the meso-tower network every hour and were used to update the mass consistent vector wind field employed in the prediction.

The measured concentration and prediction of the model at ground level at 1255 EDT are shown in Figure 8. The calculated ground level concentration of 241,000 pCi/m<sup>3</sup> at Springfield, South Carolina agrees to within 62 percent of the experimentally measured value (389,000 pCi/m<sup>3</sup>). For comparison, a Gaussian puff analytical model predicted a ground level concentration of 150,000 pCi/m<sup>3</sup>, or 39% of the experimental value.

## CONCLUSIONS

A three-dimensional method of moments numerical solution algorithm for prediction of pollutant transport within the environment has been established. The algorithm employs a quasi-Lagrangian advection scheme to minimize numerical dispersion error, with the moment distributions essentially providing sub-grid scale resolution. Cubic spline interpolation is employed to establish spatial derivatives, and the method of fractional steps is used to reduce computation time and storage requirements. Several numerical tests have validated the procedure for atmospheric predictions.

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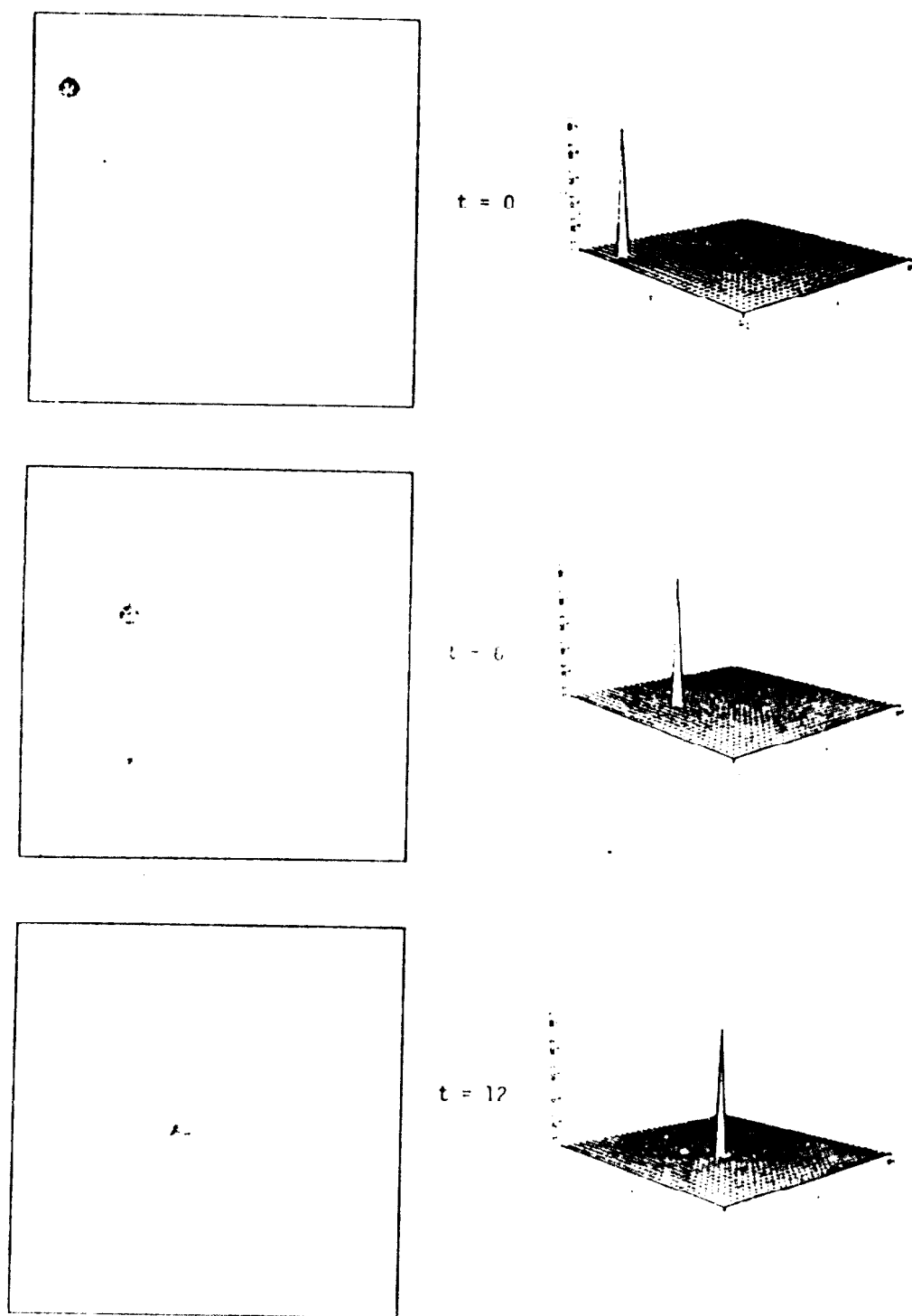
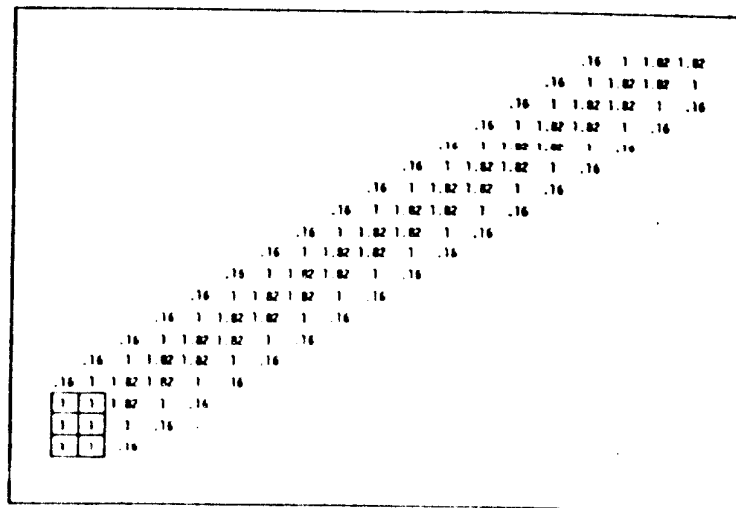
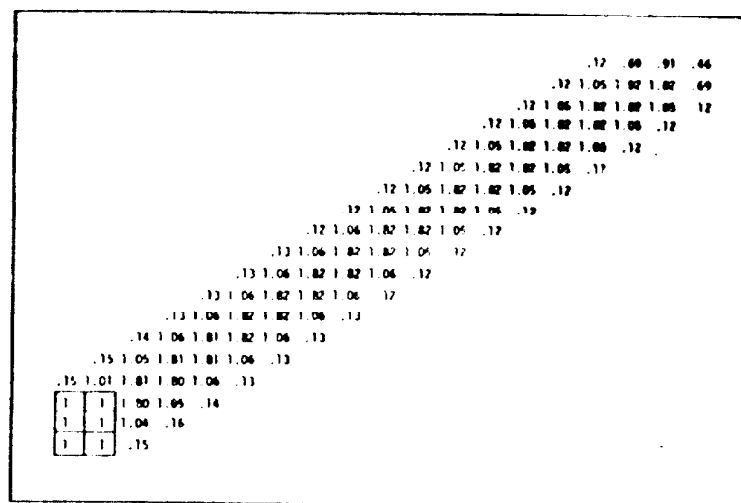


Figure 1. Advection of a Cell of Concentration ( $C = 100$ ) in Two Dimensions;  $\vec{U} = (1, -1, 0)$ ;  $\Delta X = \Delta Y = 1$ ;  $\Delta t = 0.5$



Analytical Solution (Pedersen & Prahm, 1974)



Numerical Solution

Figure 2. Advection of an Area Source ( $Q=1/\text{cell}$ );  
 $\vec{U} = (1,1,0)$ ;  $\Delta X = \Delta Y = 1$ ;  $\Delta t = 0.5$

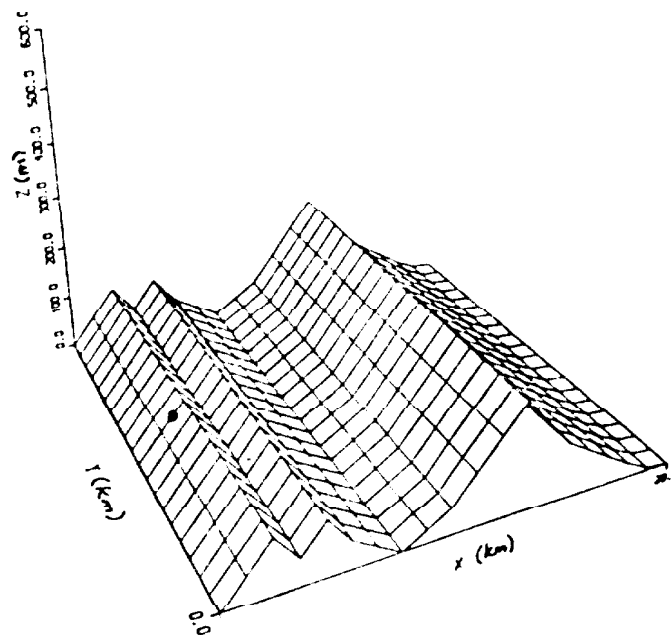
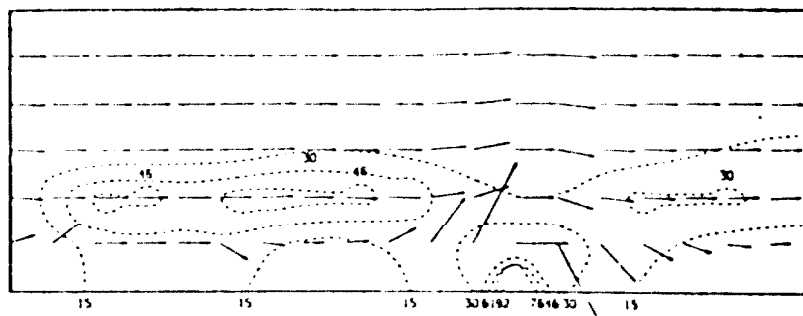
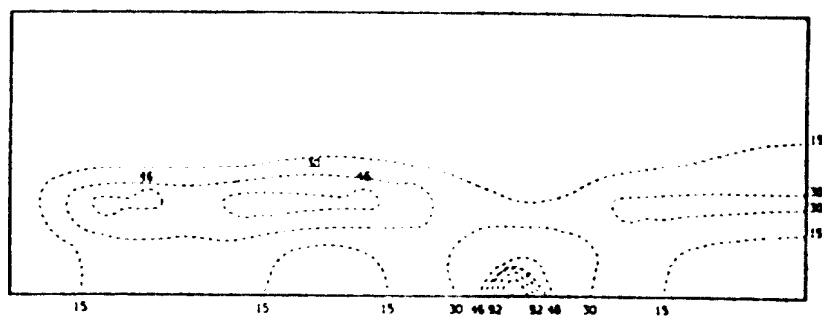


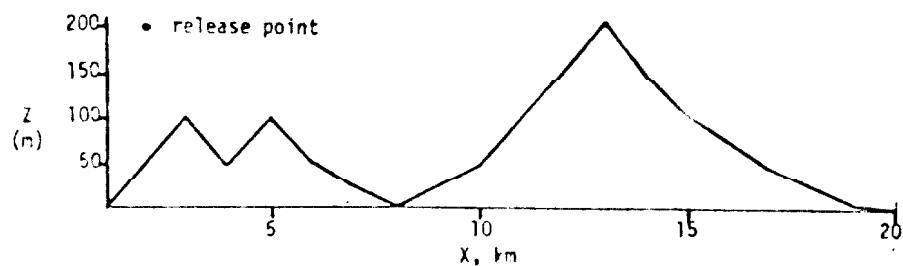
Figure 3. Topographic Surface for Continuous Elevated Emission Prediction



a.) Concentration isopleths at  $t = 1$  hour; wind vectors drawn for steady state velocities (vertical velocity component increased to enhance visualization)



b.) Concentration isopleths at  $t = 4$  hours ( $\sim$  steady state)



c.) Topography in the  $x$ - $z$  plane at  $y = \frac{1}{2}$

Figure 4. Concentration Isopleths in the  $\xi$ - $\rho$  plane at  $\eta = \frac{1}{2}$   
(Dotted Lines Denote  $C/Q$  Values in  $m^{-3}$ )

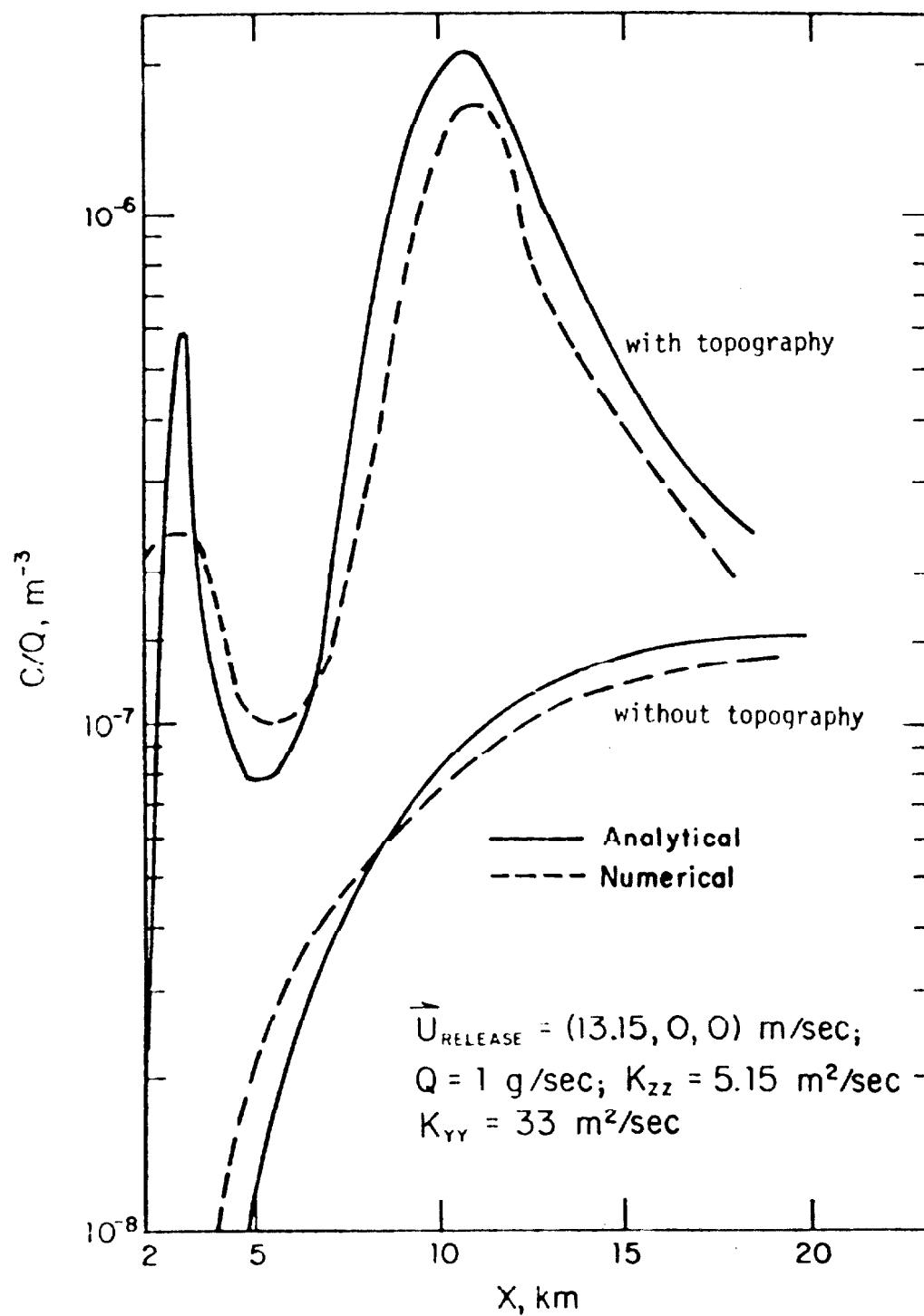


Figure 5. Ground Level Centerline C/Q Values With and Without Topography



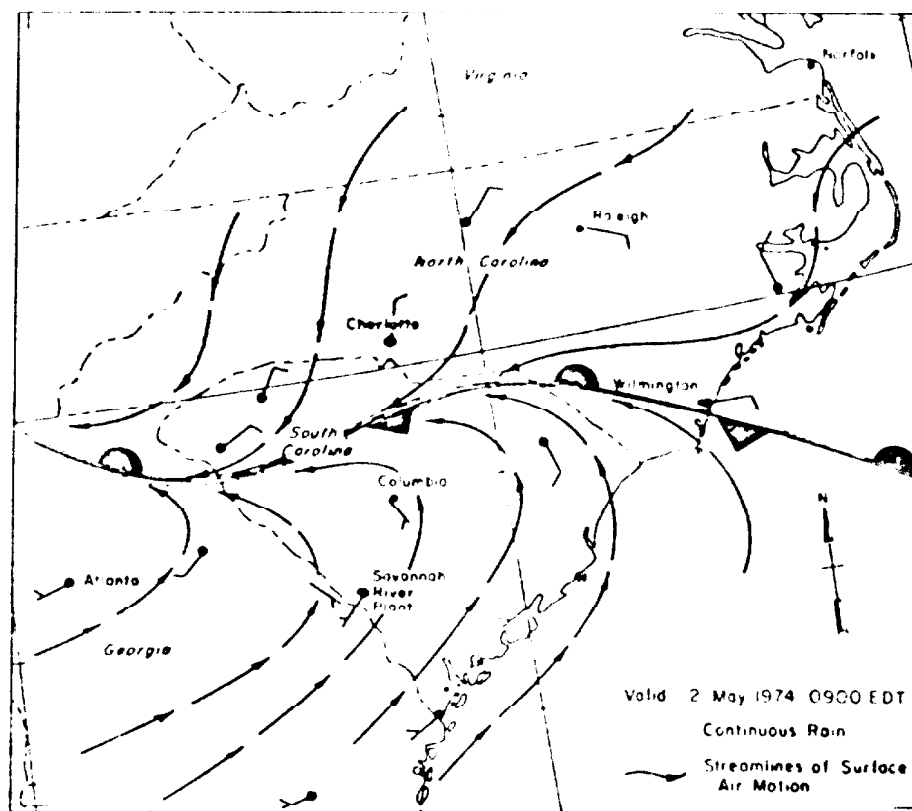


FIGURE 6. Synoptic Weather Conditions for May 2, 1974, at 0900 EDT

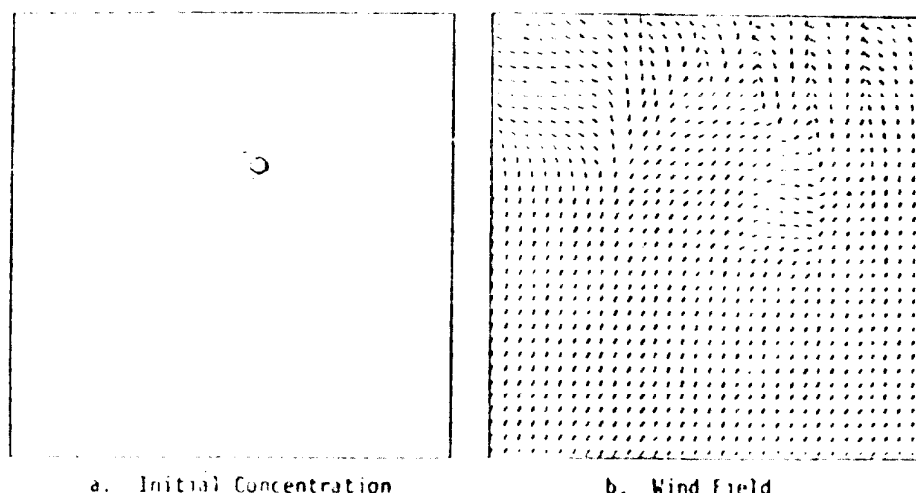


FIGURE 7. Initial Concentration Distribution and Wind Field for May 2, 1974, at  $z = 62$  m

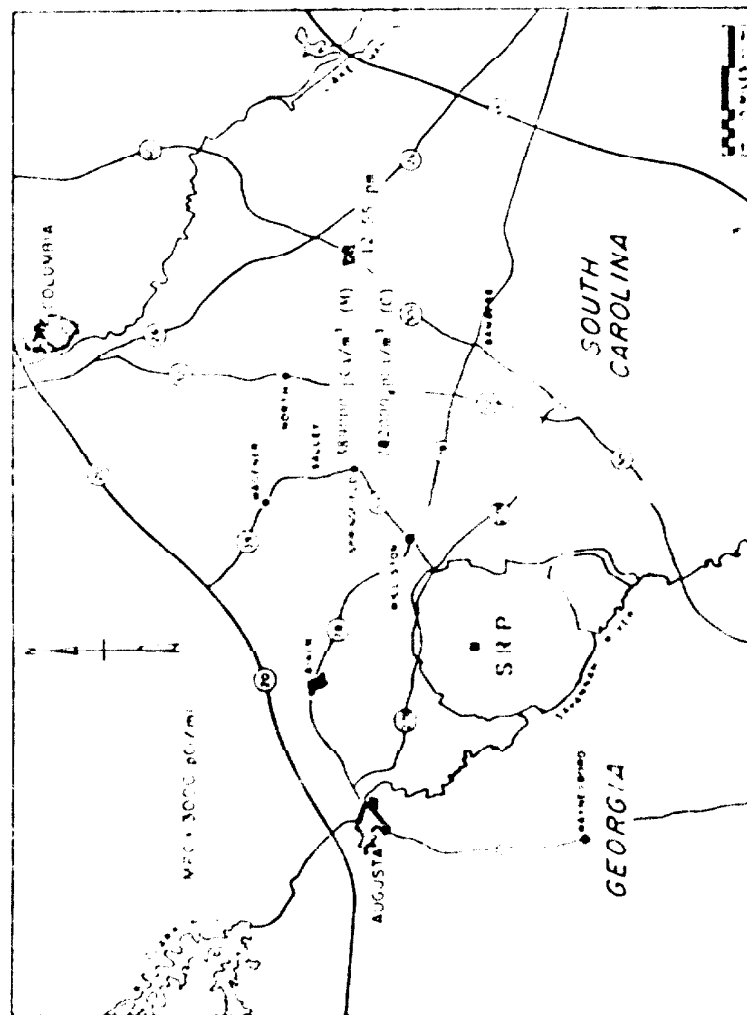


FIGURE 8. Numerical Result at Ground Level at Springfield, South Carolina for the May 2, 1974, Tritium Release. Initial release time was approximately 0800 EDT. M = measured value; C = calculated value.